



# Two Colour and Two Photon Ionization Processes in Intense Extreme-UV and Optical Laser Fields

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<http://www.ncpst.ie>

<http://www.physics.dcu.ie/~jtc>



International Workshop on EUV Sources,  
UCD, November 13-15, 2010



# Collaboration @ FLASH-DESY, Hamburg

XFEL: P. Radcliffe & M. Meyer

Paris (UPMC): R. Taieb (T) & A. Maquet (T)

PTB (Berlin): A. A. Sorokin (now at IOFFE & DESY) & M. Richter

DESY (Hamburg): K. Tiedke, S. Düsterer, W. Li, P. Juranić & J. Feldhaus

Orsay: D. Cubaynes & *D. Glijer*

Queen's University Belfast: C. Lewis, *A Delserieys, P. Orr* & H. v. d. Hart (T)

Moscow State University : A. N. Grum-Grzhimailo, E. V. Gryzlova, S. I. Strakhova

Crete: P. Lambropoulos (T)

Oulu/GSI: S. Fritzsche (T)

DCU: V. Richardson, *J. Dardis, P. Hayden, P. Hough, K. Kavanagh, T. J. Kelly, E. T. Kennedy, H. de Luna, J. P-Gutierrez* & J. T. Costello

*Thanks to AG Photon (R Treusch et al.) & AG Machine (M Yurkov et al.)*



International Workshop on EUV Sources,  
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# Collaboration @ LCLS X-ray FEL (SLAC)

MPQ/TU-Munich: A. Maier, W. Helml, W. Schweinberger & R. Kienberger

SLAC: R. Coffee, J. Hastings & J. Bozek

XFEL: P. Radcliffe, T. Tschenscher & M. Meyer

DESY (FLASH): S. Düsterer & J. Feldhaus

DESY (CFEL): I. Grguras & A. Cavalieri

Ohio (OSU): C. Roedig, G. Doumy\* & L. DiMauro

DCU: T. J. Kelly, V. Richardson, L. Nikolopoulos (T) & J. T. Costello

\*Now at Argonne.

# Some members of the current collaboration





# DCU Laser Plasma/ AMOP Group

*6 laboratory areas focussed on pulsed laser matter interactions  
(NIR – X-ray/ 30fs – 30 ns, spectroscopy/ imaging/ PLD)*

**Academic Faculty (5):** John T. Costello, Eugene T. Kennedy (Emeritus), Jean-Paul Mosnier, Lampros Nikolopoulos (T) and Paul van Kampen

**Current Postdocs (3):**

Dr. Patrick Hayden, Dr. Sateesh Krishnamurthy  
and Dr. Subhash Singh

**Funded by:**

SFI - Frontiers and Investigator  
HEA – PRTL (Kit)  
IRCSET (People)  
EU - Marie Curie (People)

**Current PhD students (10):** Jack Connolly, Leanne Doughty, Brian Doohan, Colm Fallon, Eanna MacCarthy, Mossy Kelly, Vincent Richardson, Jiang Xi, Damien Middleton, Cathal O'Broin

**Visiting Students:**

Ricarda Laasch (Univ. Hamburg) and Nadia Gambino (Univ. Catania)

**Interns:** Deirdre Cogan, Michael Creagh, Adrian Maguire and Florian Wruck

**DCU International Visiting Fellow:** Prof. Sivanandan Harilal (Purdue)



International Workshop on EUV Sources,  
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# Outline of Talk - Web Version

1. EUV FEL Fundamentals
2. Two Colour Femtosecond Pump-Probe Setup at the FLASH FEL
3. Two Colour (Above Threshold) Ionization  
*EUV-Optical Pulse Synchronisation at FLASH*  
*Atomic Dichroism in He at FLASH*  
*Long (> 100 fs) Pulse Width Measurements at the LCLS*  
*Ultrashort (<~ fs) Pulse Width Measurements at the LCLS*
4. One Colour - 2 Photon Ionization  
*Case Study 1: 2-Photon 4d-ionization of Xe (ATI)*  
*Case Study 2: Resonant 2-Photon 3d-4d excitation in Kr*
5. Some conclusions

# Outline of Talk - 15 Min Version

1. FLASH Specifications at 13.5 nm
2. Two Colour Femtosecond Pump-Probe Setup at the FLASH FEL
3. Two Colour (Above Threshold) Ionization  
*EUV-Optical Pulse Synchronisation*  
Long ( $> 100$  fs) Pulse Width Measurements at the LCLS  
*Ultrashort ( $< \sim$  fs) Pulse Width Measurements at the LCLS*
4. One Colour - 2 Photon Ionization  
*Case Study 1: 2-Photon 4d-Ionization of Xe (ATI)*
5. Some conclusions

# Ireland & EUV & X-Ray FELs

*Collaboration grew out of EU RTD Project:  
HRPI-CT-1999-50009*

**Title: “X-Ray FEL Pump Probe Facility”**

**Partners:**

**Orsay, DCU, Lund, MBI, BESSY & DESY**



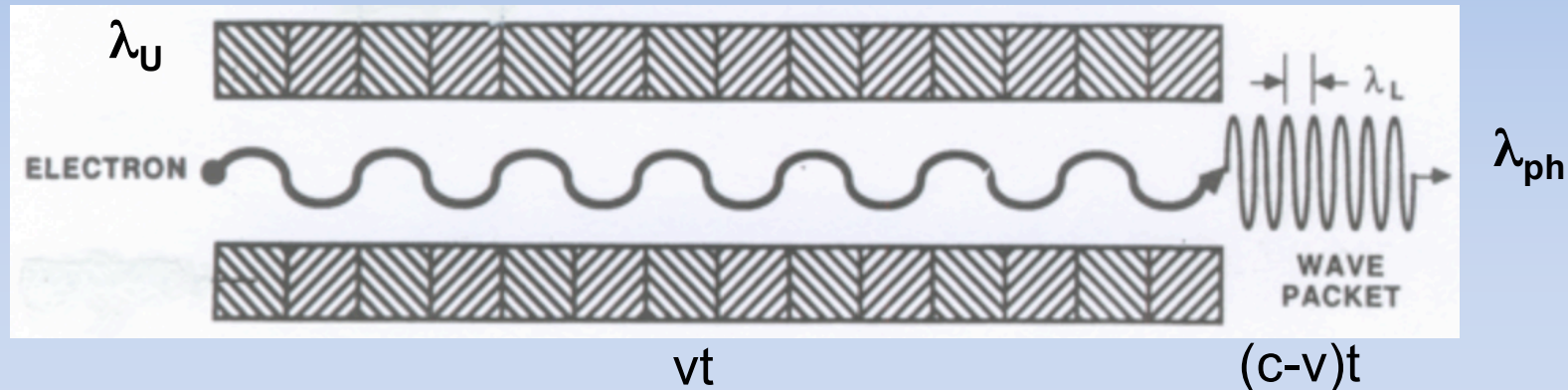
International Workshop on EUV Sources,  
UCD, November 13-15, 2010





# 1. SASE-FELs - Fundamental Principle

**SASE FEL Requirements:** Mono-energetic relativistic electron bunch of very high charge density  
And low emittance with an ultra-precise long magnetic undulator in perfect alignment



$$N_u \lambda_u = vt$$

$$N_u \lambda_{ph} = (c-v)t \Rightarrow \lambda_{ph} \sim \lambda_u (c-v)/v \sim \lambda_u / 2\gamma^2$$

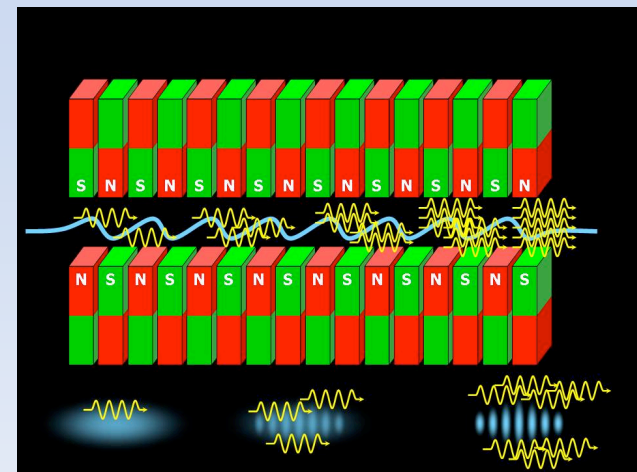
1GeV machine  $\gamma \sim 2000$

$\lambda_u \sim 2.7 \text{ cm} / \lambda_{laser} \sim 6 \text{ nm}$

$$\lambda = \lambda_u (1 + K^2/2) / 2\gamma^2$$

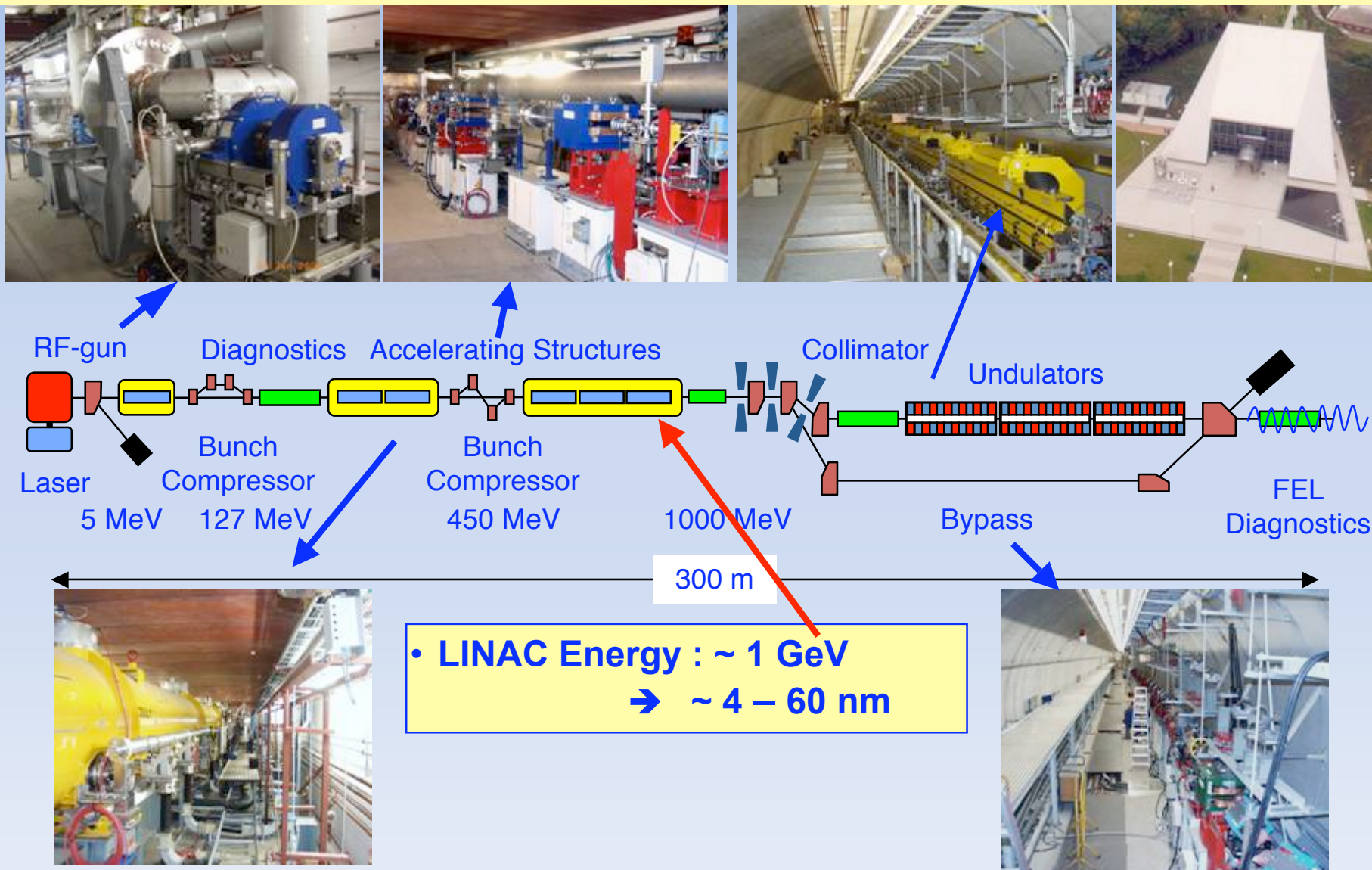
$$K = eB \lambda_u / 2\pi mc \quad \gamma = E/mc^2$$

**Electron bunch slips behind lightwave by  $\lambda$  per undulator period - *microbunching***



<http://feldfrei.wordpress.com>

# (DESY-Hamburg) 'FLASH' FEL Layout





# nature photonics

VOL. 1 NO. 6 JUNE 2007  
www.nature.com/naturephotonics

Entering the  
water window

QUANTUM CRYPTOGRAPHY  
Going the distance

METROLOGY  
Vapour cells on a chip

START-UPS  
Entrepreneur's story

## **FLASH:** **Key Performance Indicators**

**Wavelength range (fundamental):**

4 - 60 nm (2010)

**FEL harmonics (@13.7 nm):**

3 rd : 4.6 nm (270 eV)

5 rd : 2.7 nm (450 eV)

**Spectral width (FWHM):**

0.5-1 %

**Pulse energy:**

up to 50  $\mu\text{J}$  (average),

120  $\mu\text{J}$  (peak)

**Pulse duration (FWHM):**

10-20 fs

**Peak power (fundamental):**

Few GW

**Average power (fundamental):**

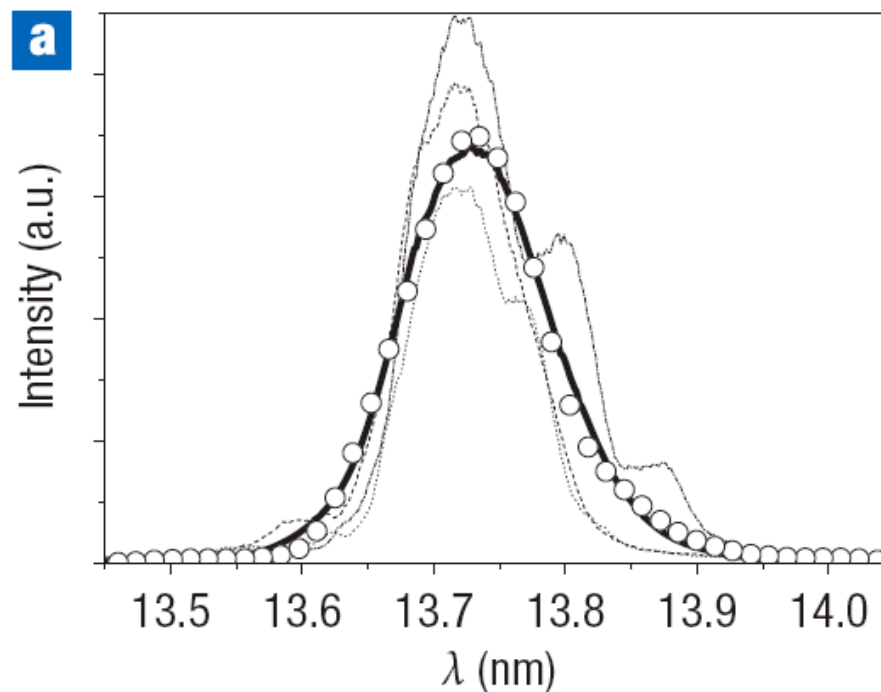
0.5 W (up to 10000 pulses /sec)

**Photons per pulse:**

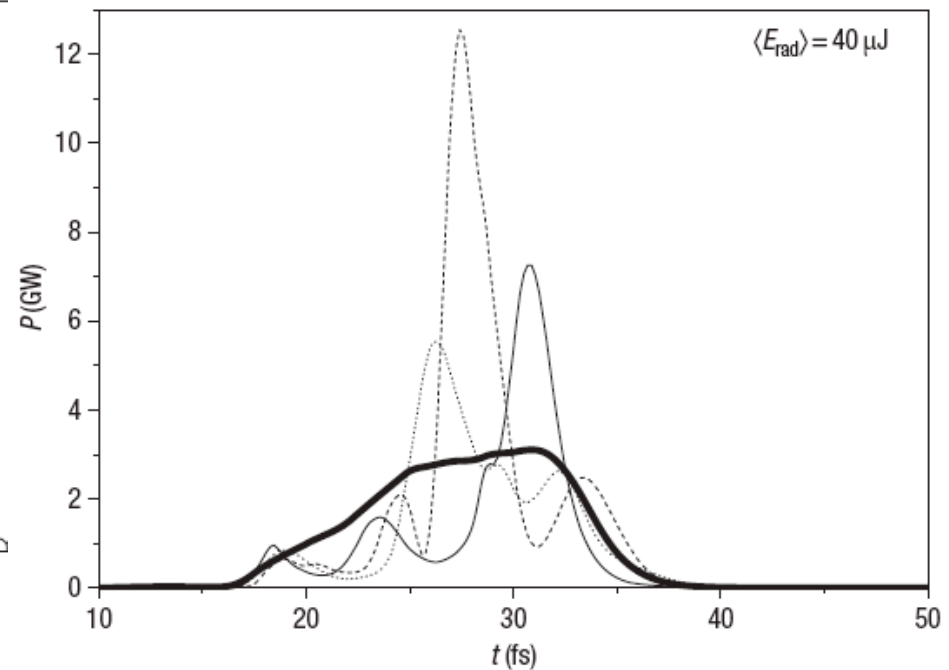
$\sim 10^{13}$

# O/P Profile and Spectra @ 13.5 nm

FEL output builds up from spontaneous emission (photon noise) => **SASE** – ‘**Self Amplified Spontaneous Emission**’ => **Fluctuations in beam profile, pointing stability, intensity, spectral distribution and pulse duration !!**  
*Spatially coherent only - ‘Seeded FEL projects at FLASH & LCLS’*



Spectral Fluctuations



Temporal Fluctuations



# (Currently) Operating EUV / X-ray SASE FEL Facilities Worldwide - Parameters

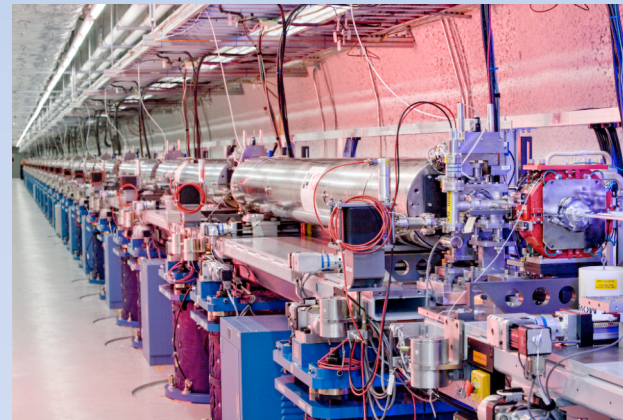
**1. FLASH:** 30 – 300 eV/10 Hz (1 – 1000 micropulses/bunch) - 10uJ/10fs/micropulse (CW RF)

**2. LCLS:**

1 nC bunch: 0.8 – 12 keV  
(60 Hz)/ 2.5 mJ/250 fs

10 pC bunch: 0.8 – 12 keV  
(60Hz)/50 uJ/ 1 – 4 fs

**3. Spring-8 (SCSS):** 20 - 22 eV  
(60Hz)/ 50 uJ/ 100 fs



# USPs of XFELs in AMO Physics ?

- *Ultra-dilute* targets
- *Photo*-processes with *ultralow cross-sections*
- *Pump and probe* experiments (EUV + EUV *or* EUV + Opt.)
- *Single shot* measurements
- *Few-photon* single and multiple *ionization processes*

NB1: Makes *inner-shell electrons* *key actors* in non-linear processes for the first time

NB2: Re-asserts *primacy of the photon* over field effects !

# EUV / XFEL AMO Physics Reviews

## 1. Photoionization Experiments with the Ultrafast XUV Laser FLASH

J. T. Costello, *J. Phys. Conf. Series* **88** Art No. 012057 (2007)

## 2. Experiments at FLASH

C. Bostedt, H. N. Chapman, J. T. Costello, J. R. Crespo Lopez-Urrutia, S. Duesterer, S. W. Epp, J. Feldhaus, A. Foehlich, M. Meyer, T. Mšller, R. Moshhammer, M. Richter, K. Sokolowski-Tinten, A. Sorokin, K. Tiedtke, J. Ullrich and W. Wurth,  
*Nucl. Inst. Meth. in Res. A* **601** pp108-122 (2009)

## 3. Non-linear processes in the interaction of atoms and molecules with intense EUV and X-ray fields from SASE free electron lasers (FELs)

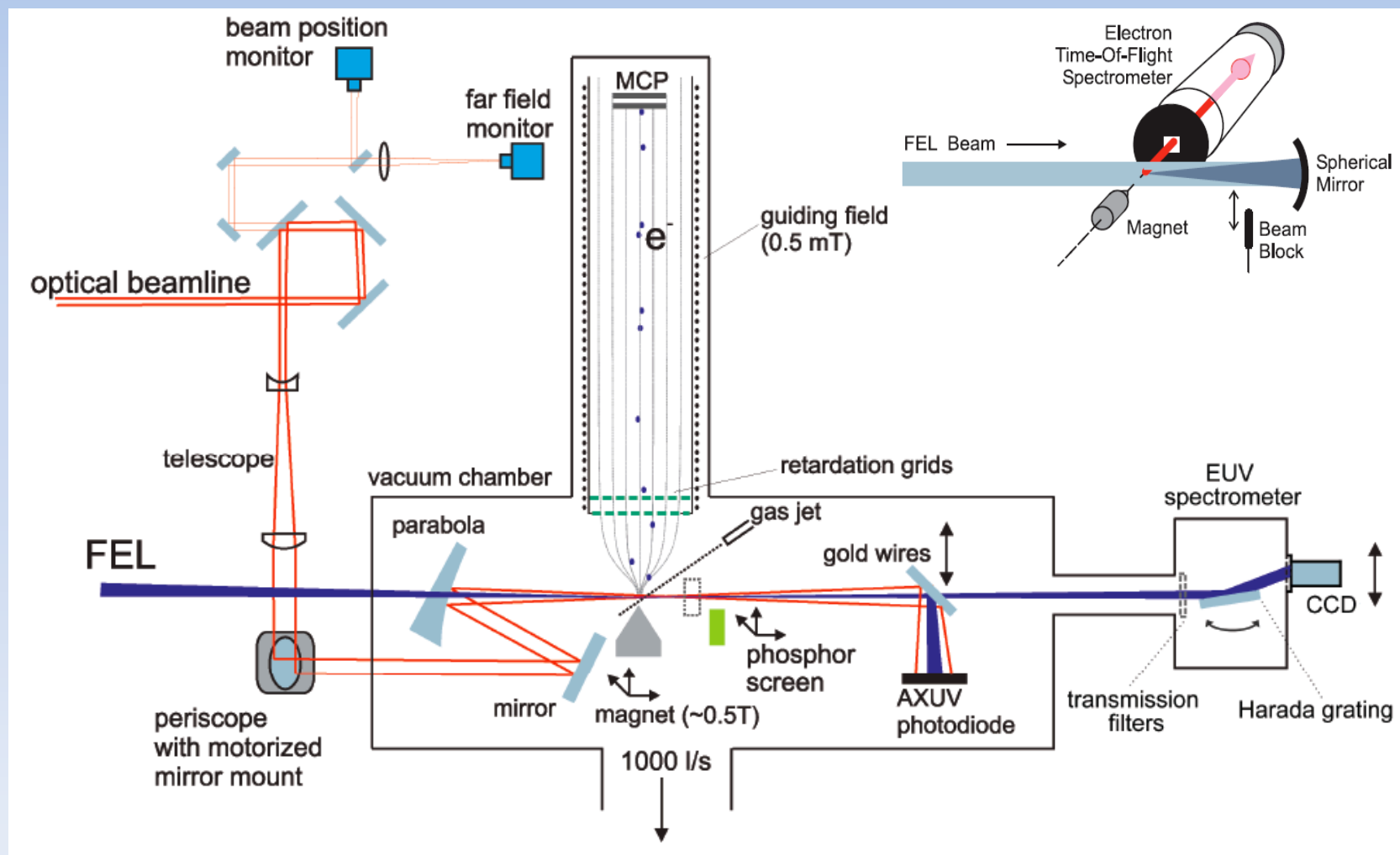
N. Berrah, J. Bozek, J. T. Costello, S. Duesterer, L. Fang, J. Feldhaus, H. Fukuzawa, M. Hoener, Y. H. Jiang, P. Johnsson, E. T. Kennedy, M. Meyer, R. Moshhammer, P. Radcliffe, M. Richter, A. Rouzee, A. Rudenko, A. Sorokin, K. Tiedtke, K. Ueda, J. Ullrich and M. J. J. Vrakking,  
*Journal of Modern Optics* **57** pp 1015-1040 (2010)

## 4. Two-colour experiments in the gas phase

M. Meyer , J. T. Costello , S. Düsterer , W. B. Li and P. Radcliffe  
*J. Phys. B: At. Mol. Opt. Phys.* **43** Art No. 194006 (2010)

# 2. Photoelectron Spectroscopy Setup

## Experimental Layout at FLASH - (EU-RTD)





# Part 3. Atoms in Intense EUV (X-ray) + Optical (800 nm/ 2000 nm) fields

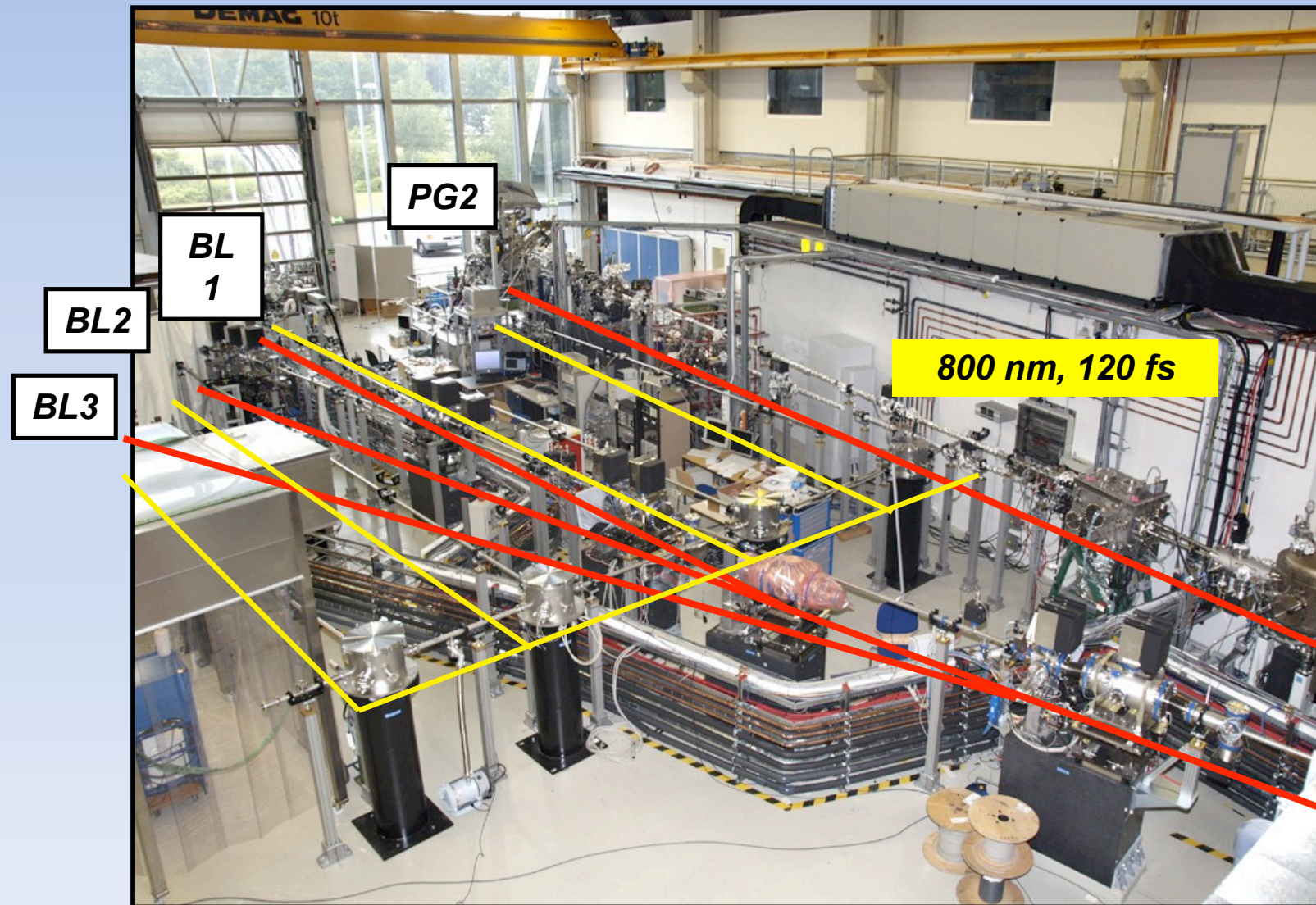
*Case 1. EUV-Optical Pulse Synchronisation*

*Case 2. (FLASH) - Dichroism in He  $\sim 13.5$  nm*

*Case 3. Long ( $> 100$  fs) Pulse Width  
Measurements at the LCLS*

*Case 4. Ultrashort ( $< \sim$  fs) Pulse Width  
Measurements at the LCLS*

# FLASH NIR/UV and XUV Beam Layout

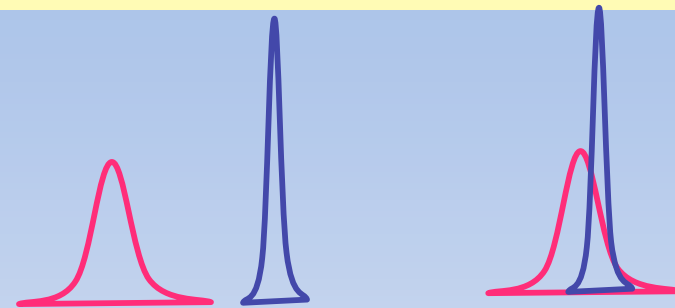


# Two colour ATI/ Laser Assisted PES

Superposition of visible and XUV pulses in a noble gas jet

*Schins et al. PRL 73, 2180 (1994)*

$$A + \hbar\omega_{XUV} \rightarrow A^+ + e^-(T_{KE}) \pm n\hbar\omega_L \rightarrow A^+ + e^-(T_{KE} \pm n\hbar\omega_L)$$

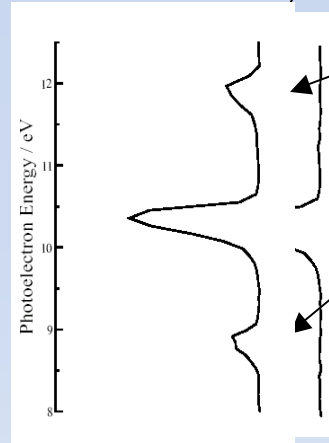


Electron Spectrometer

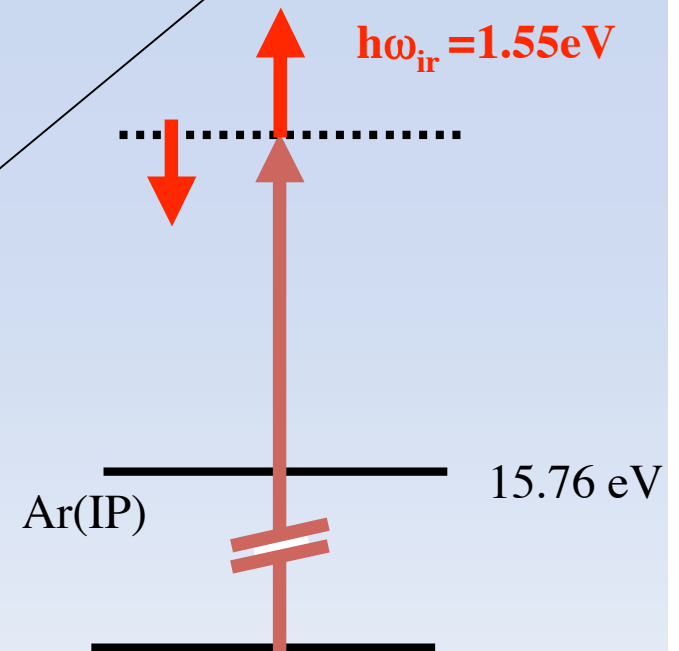


NIR (800 nm) fs laser pulse

Gas Jet



Sideband intensity  
very sensitive to XUV-  
IR pulse area overlap.  
- Cross Correlation...



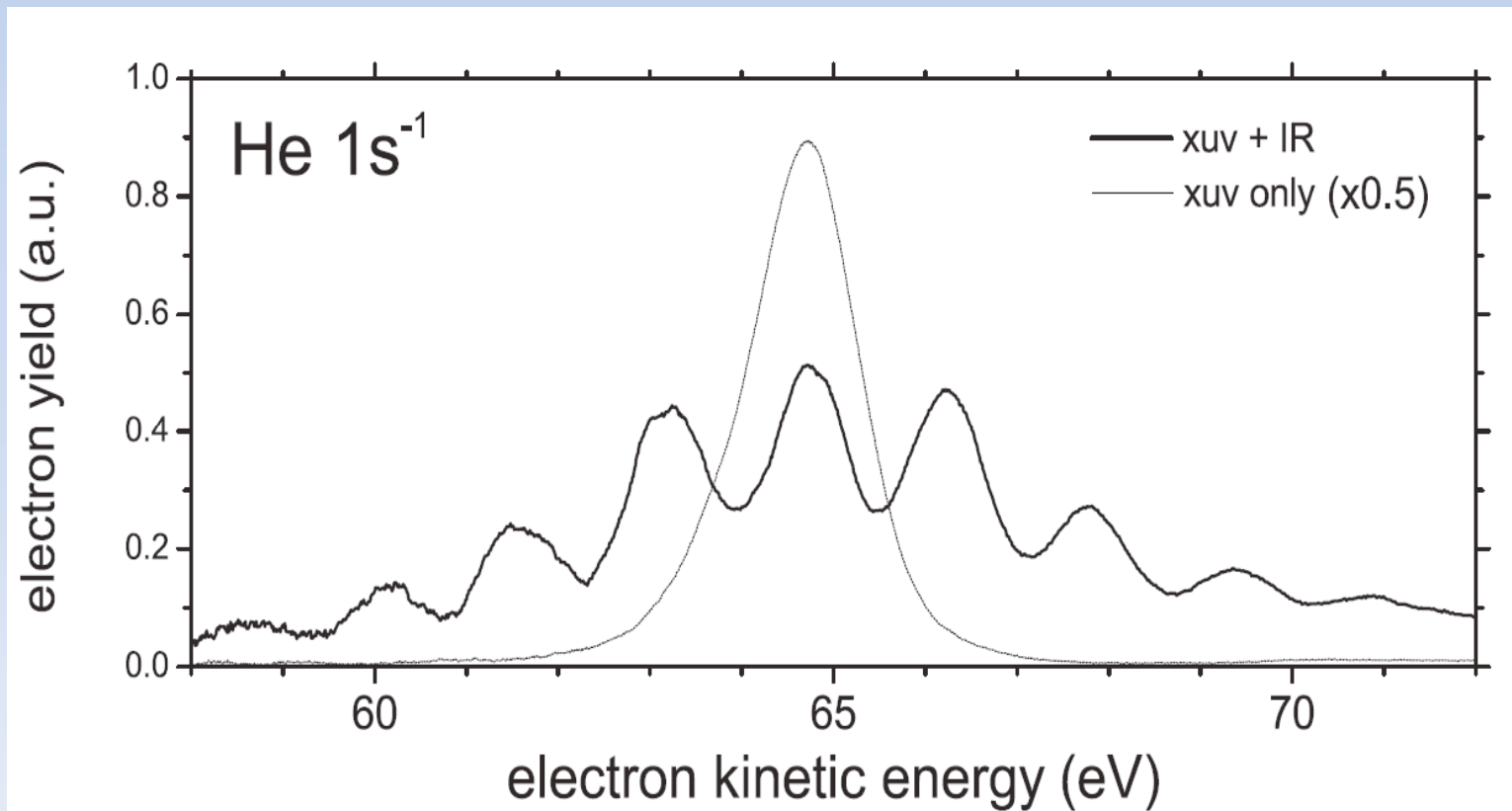
# Two colour ATI/ Laser Assisted PES

FLASH: 13.7 nm,

10-30 fs, 20μJ

Optical Laser: 800nm,

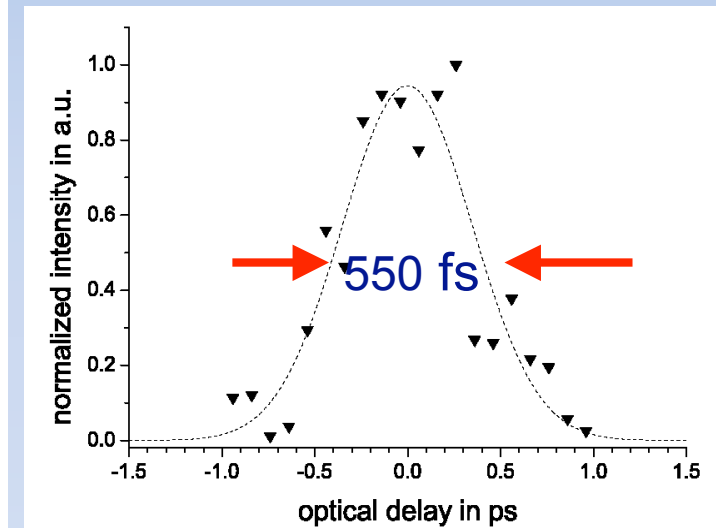
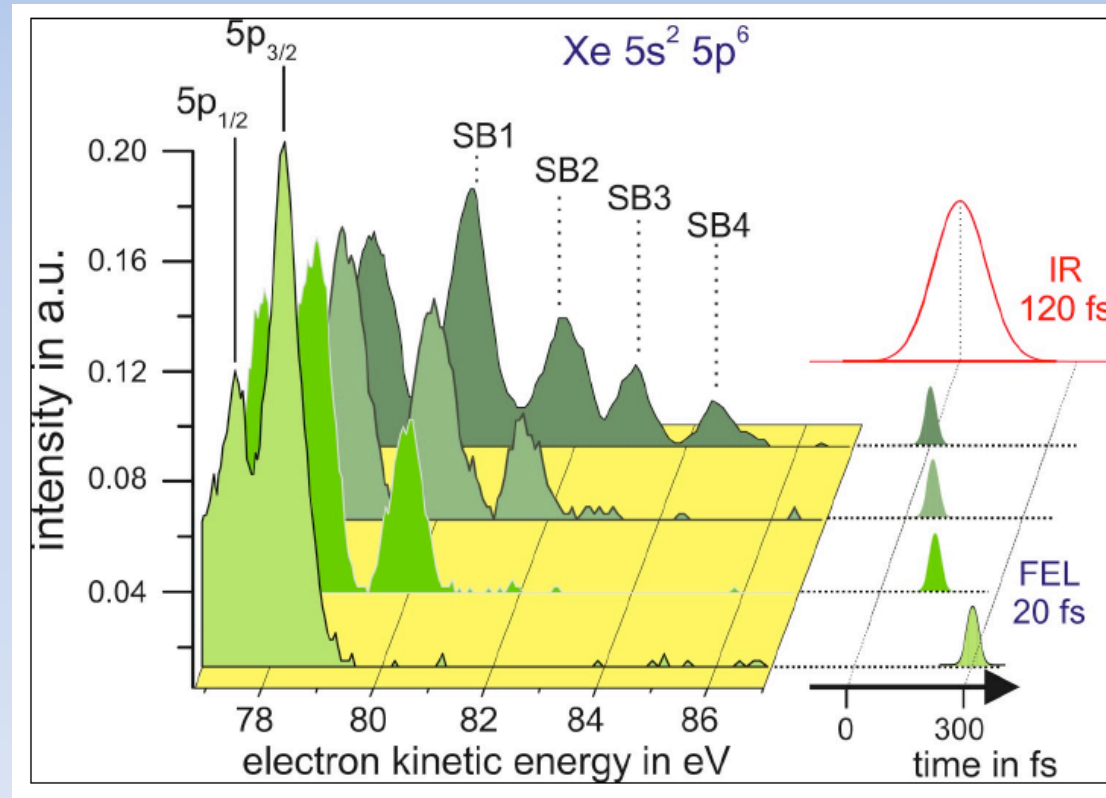
120 fs, 400μJ,  $5 \times 10^{13} \text{ W/cm}^2$





# Two colour ATI/ Laser Assisted PES

Sideband number/intensity depend strongly on XUV/NIR overlap  $\Rightarrow$  by comparison with theory *we are able to determine relative time delay to better than 100 fs*



## 1. New ultrafast XUV-modulated optical-reflectivity methods

C. Gahl et al., Nature Photonics **2** 165-169 (2008)

T. Maltezopoulos et al., New J Phys **10** Art. No. 033026 (2008)

## 2. 'TEO'

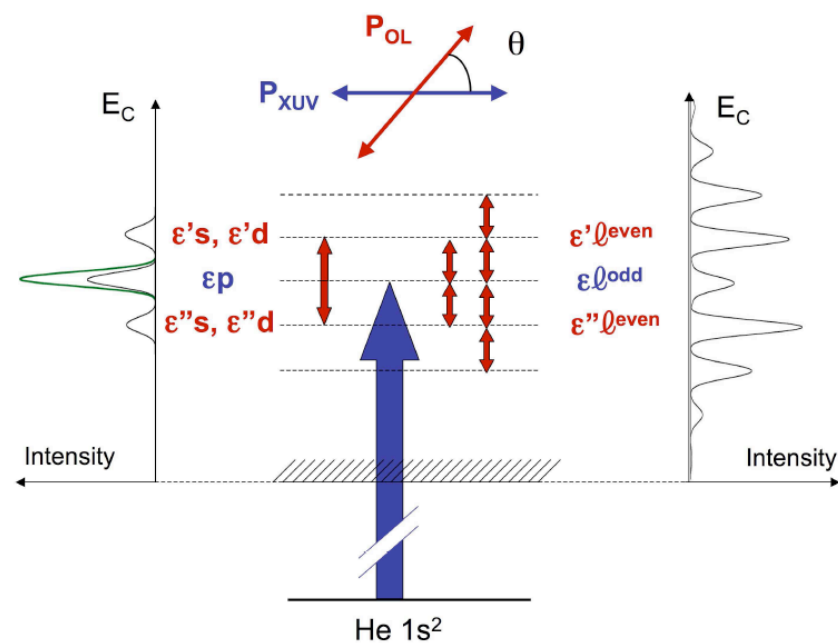
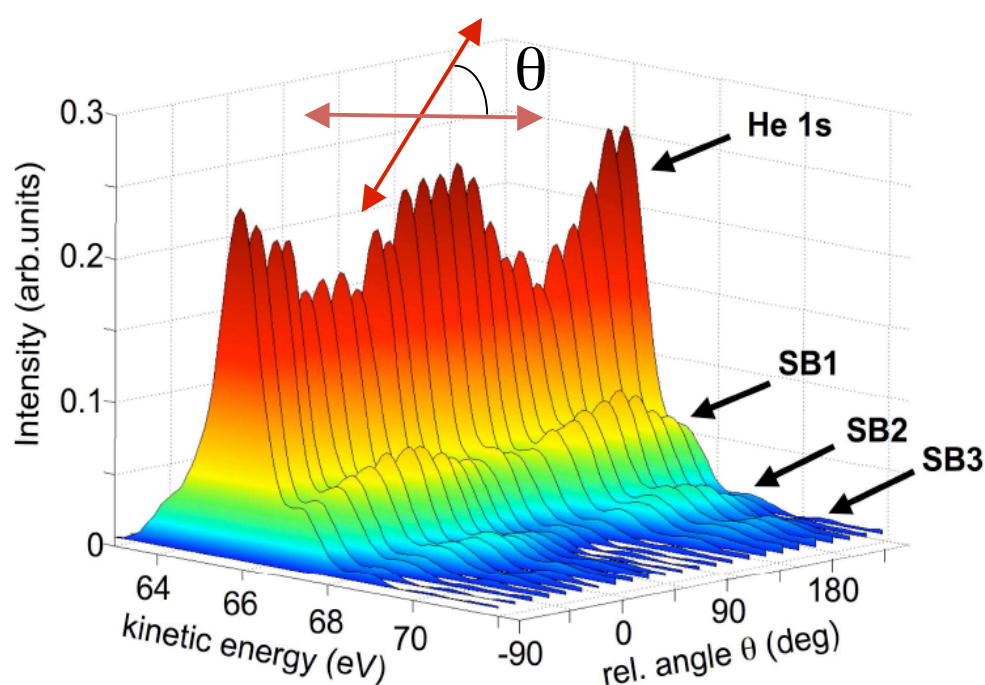
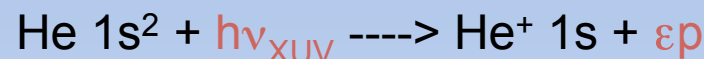
A. Azima et al., APL, **94** 144102 (2009)

# Atomic Dichroism in Two Colour ATI - He

## Strong Polarisation Dependence of Sidebands (Low Field)

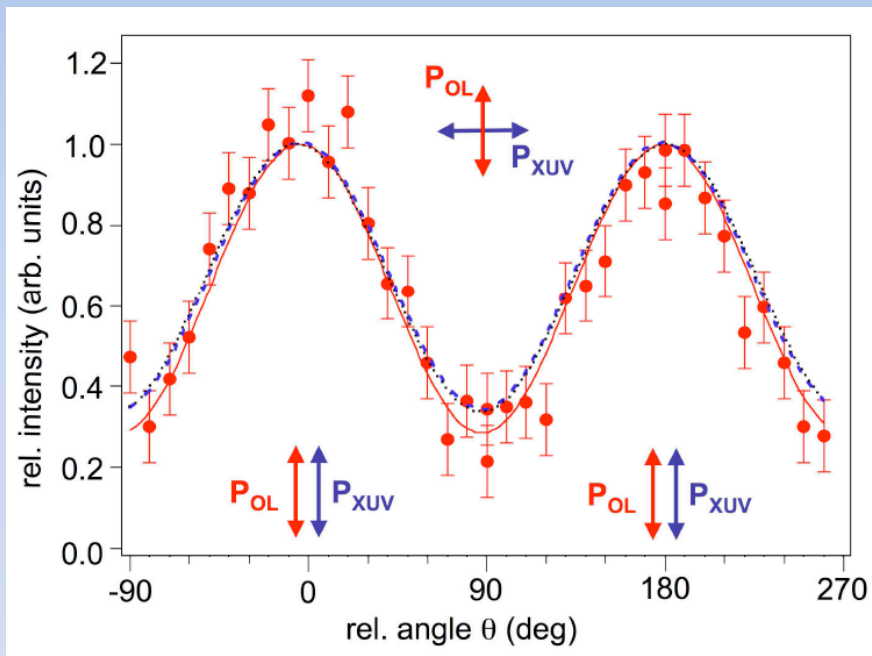
FLASH: 13.7 nm, 10-20 fs, 20μJ

OL: 800nm, 4ps, 400μJ,  $6 \times 10^{11}$  W/cm<sup>2</sup>



# Atomic Dichroism in Two Colour ATI - He

## Low Optical Laser Field

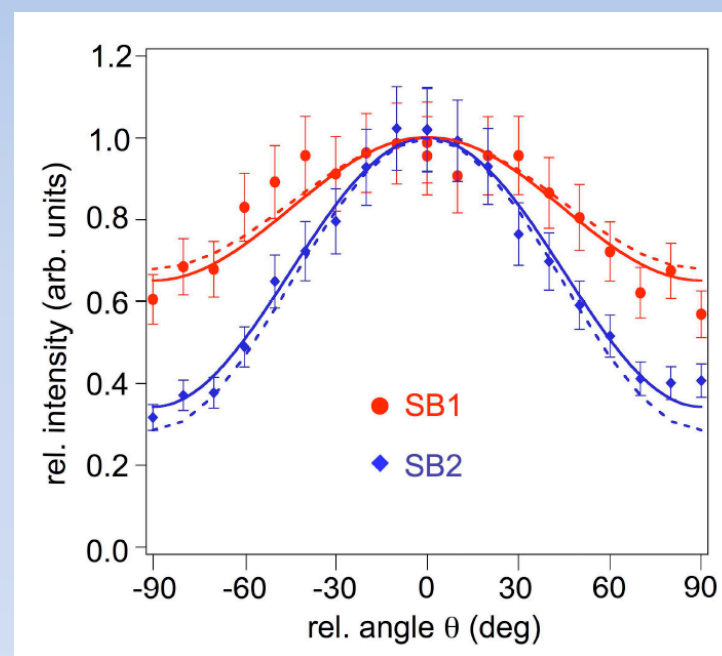


$$\sigma(\theta) = 3S_d + (5S_s + S_d) \cos^2\theta$$

$$S_s/S_d = 1.25 \pm 0.3$$

**P. Theory:** A Grum-Grzhimailo et al.

## High Optical Laser Field



$$\left( \frac{d\sigma^{(n)}}{d\theta} \right) = J_n^2(\vec{\alpha} \cdot \vec{K}) \left( \frac{d\sigma^{(0)}}{d\theta} \right)_{\vec{E}_k}$$

$$S_{He}^{(n)} \propto \int_0^\pi \sin\theta ( \cos^2\theta ) J_n^2(\alpha_0 k_n \cos\theta) d\theta$$

**SPA:** A Maquet/ R Taieb

# Two Colour X-ray + NIR Expts @ LCLS

**Collaboration:** R. Kienberger, J. Bozek, A. Cavalieri, R. Coffee, J. Costello, S. Duesterer, M. Meyer, L. DiMauro & T. Tschentscher -  
*MPQ, SLAC, DESY-CFEL, DCU, DESY-F:ASH, XFEL & Ohio State*

## *Few Femtosecond Photo and Auger Electron Dynamics in Strong Optical Fields*

### **Idea.**

LCLS has a 'low bunch current mode' which allows it to deliver ultrashort pulses of duration: 1 - 4 fs.....

So we are using the Single Shot Atomic Streak Camera or **SSASC** technique\* to measure it.

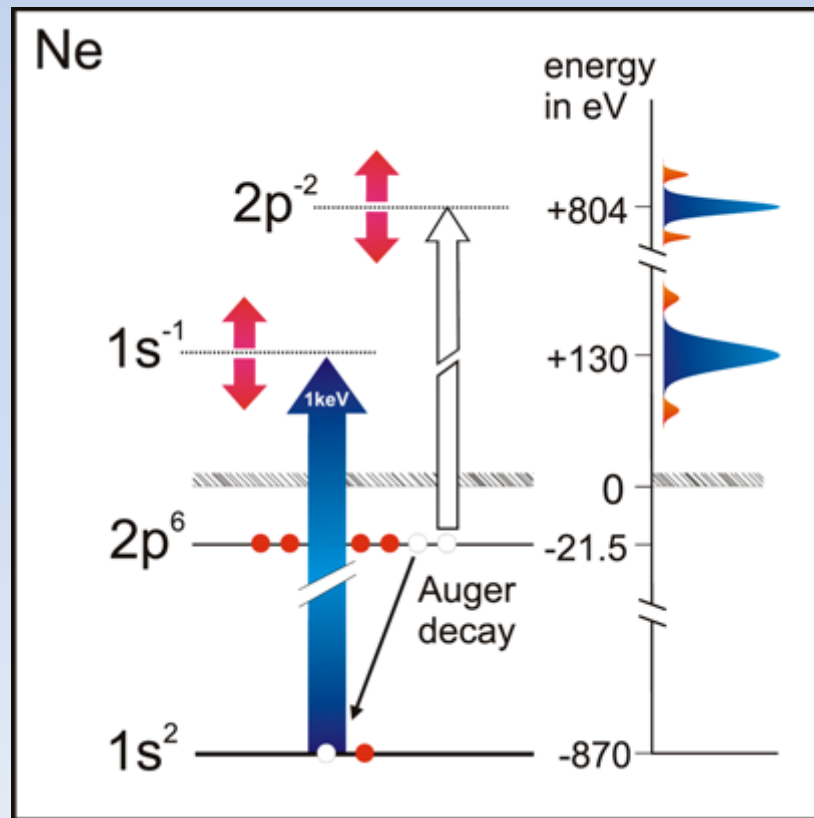
\* *R. Kienberger et al., J. Mod. Opt* **52** 261-275 (2005)

# Two Colour X-ray + NIR Expts @ LCLS

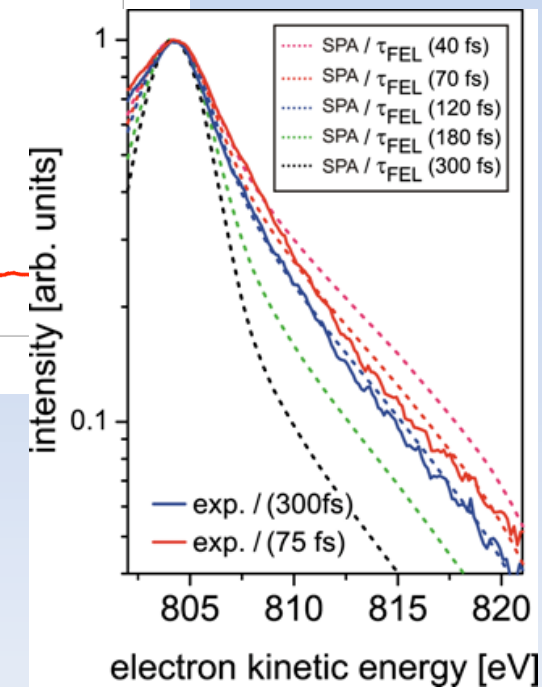
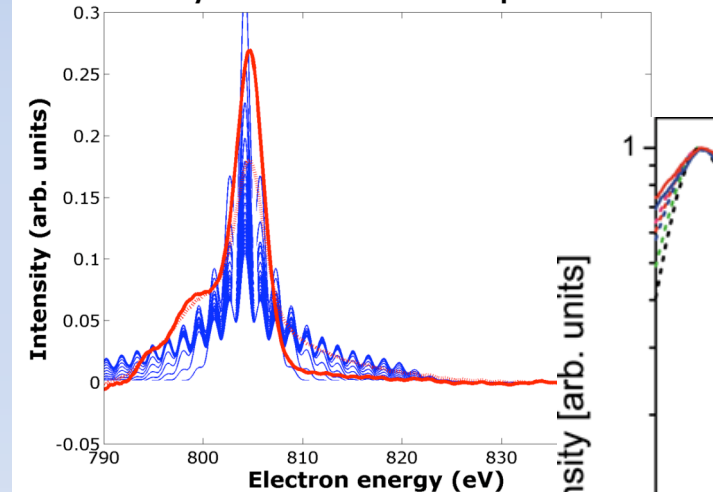
First test experiment at LCLS - LONG X-RAY PULSES - sideband generation.

**Target:** Ne, **LCLS:** 1 keV & ~50 -300 fs, **Laser:** 800 nm/ ~1 mJ & < 100 fs

**Key Message:** 300 fs (from FEL parameters) = 120 +/- 20 fs optical from SB analysis..  
70 fs (from FEL parameters) = ≤ 70 fs optical from SB analysis..



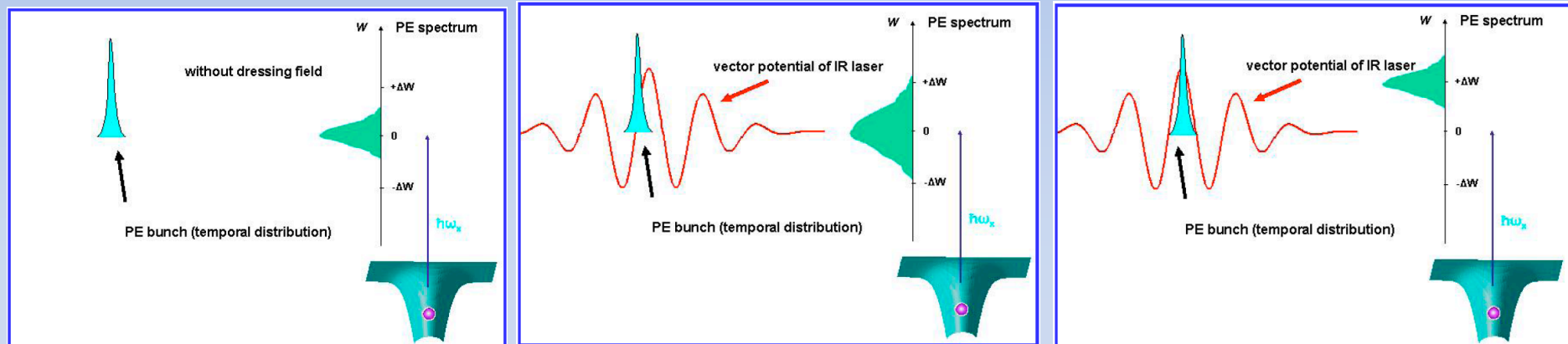
for IR intensity 0 -  $1 \times 10^{12} \text{ Wcm}^{-2}$  in steps of  $5 \times 10^{10} \text{ Wcm}^{-2}$



# Two Colour X-ray + NIR Expts @ LCLS

**Single Shot Atomic Streak Camera – SSASC => few fs pulse widths**

**Target:** Neon, **LCLS:** >850 eV, ~1 - 4 fs, **Laser:** OPA (2000 nm, ~ 7 fs),



a. Without dressing field => unshifted, no broadening.....

b. With dressing field (zero crossing) => unshifted, broadened.....

c. With dressing field (peak value) => shifted, not broadened.....

1. Electron bunch must replicate ultrashort X-ray pulse.....Neon is the Photocathode !!
2. Electron bunch duration must be shorter than one half cycle of the OPA dressing field (~ 3fs).....
3. Photoelectron energy shift follows the electric field of the IR dressing laser.....
4. In the zero field crossing case the electron pulse is streaked in both directions resulting in a broadened but unshifted electron line - the electron linewidth will depend on the electron (X-ray) bunch length (case 'b')
5. On the other hand, if the electron bunch falls on the carrier peak, all parts of the bunch 'feel' approximately the same dressing field, ergo the electron bunch will be shifted by a constant energy (case 'c')
6. Postprocessing - retrieve zero crossing cases to determine LCLS pulse width distribution (< 1 to 4 fs ?)



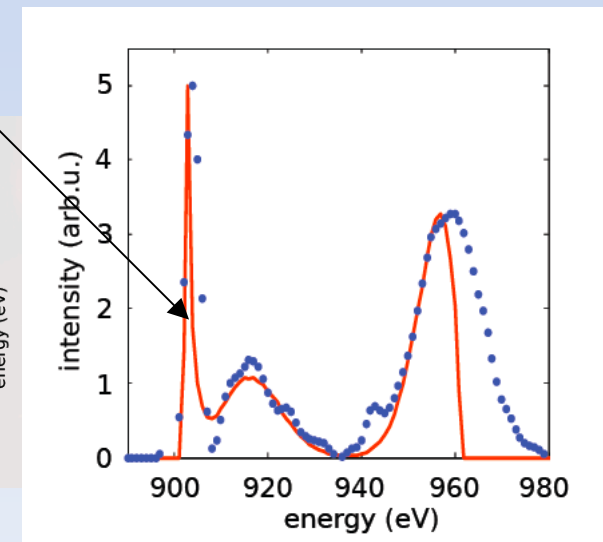
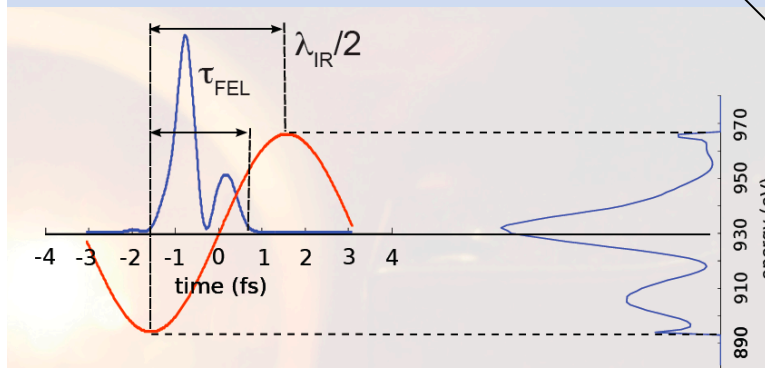
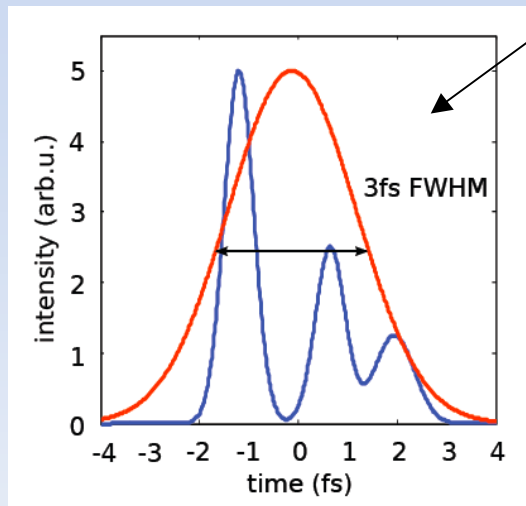
# Two Colour X-ray + NIR Expts @ LCLS

*Few fs mode - under analysis.....*

Process.  $\text{Ne} + h\nu (1.8 \text{ keV}) \rightarrow \text{Ne}^+ (1s^{-1}) + e^- + I_L (10^{14} \text{ W.cm}^{-2})$

Essentially mapping time (fs) to energy in (eV) allows one to measure X-ray (and EUV) pulse widths to attosecond accuracy provided the *X-ray (EUV) pulse width is less than one one half cycle of the optical laser in duration !!*

3fs case - simulation and experiment.....



# Part 4. Two Photon Ionization (TPI) of atoms in an Intense Field

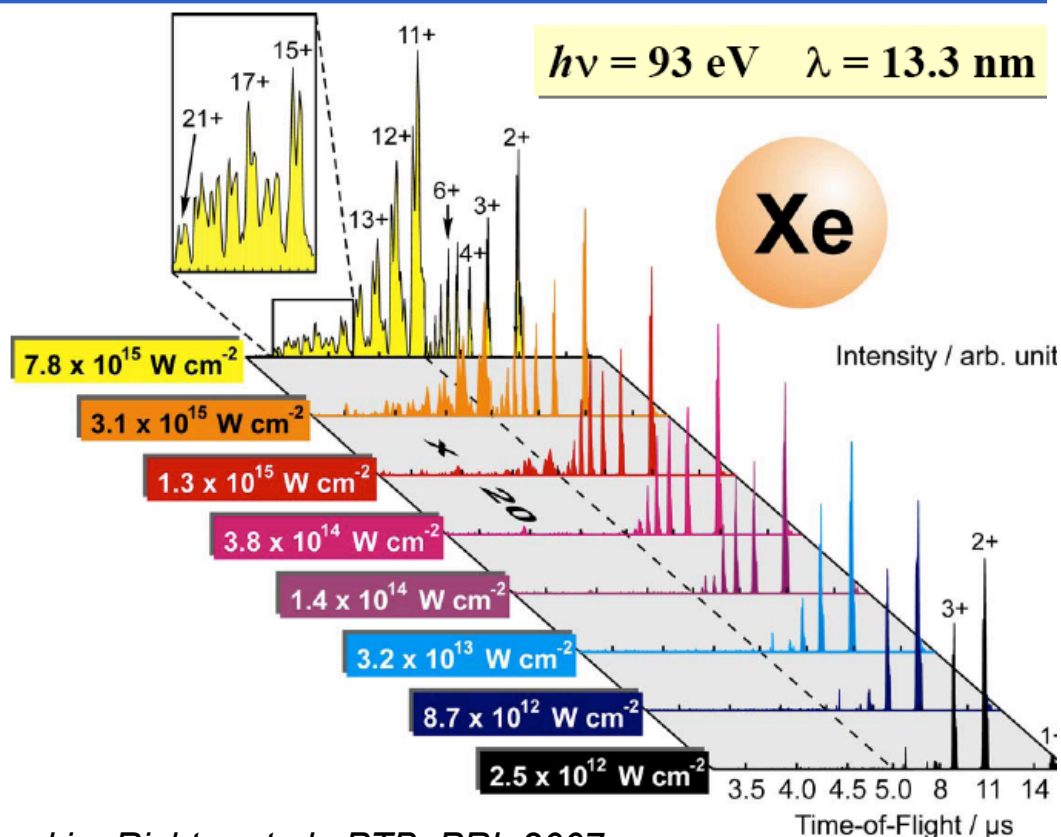
*Case 1. Non-Resonant: Xe @  $h\nu = 93$  eV*

*Case 2. Resonant: Kr @  $h\nu = 46$  eV (3d-4d)*

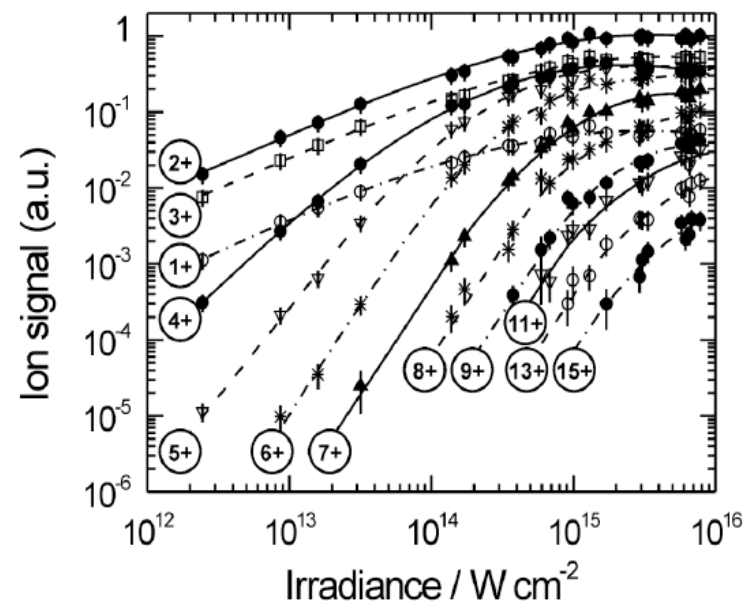
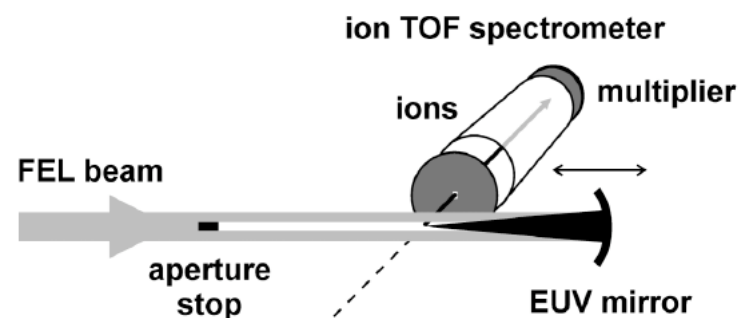
# Motivation - Xe in intense EUV fields

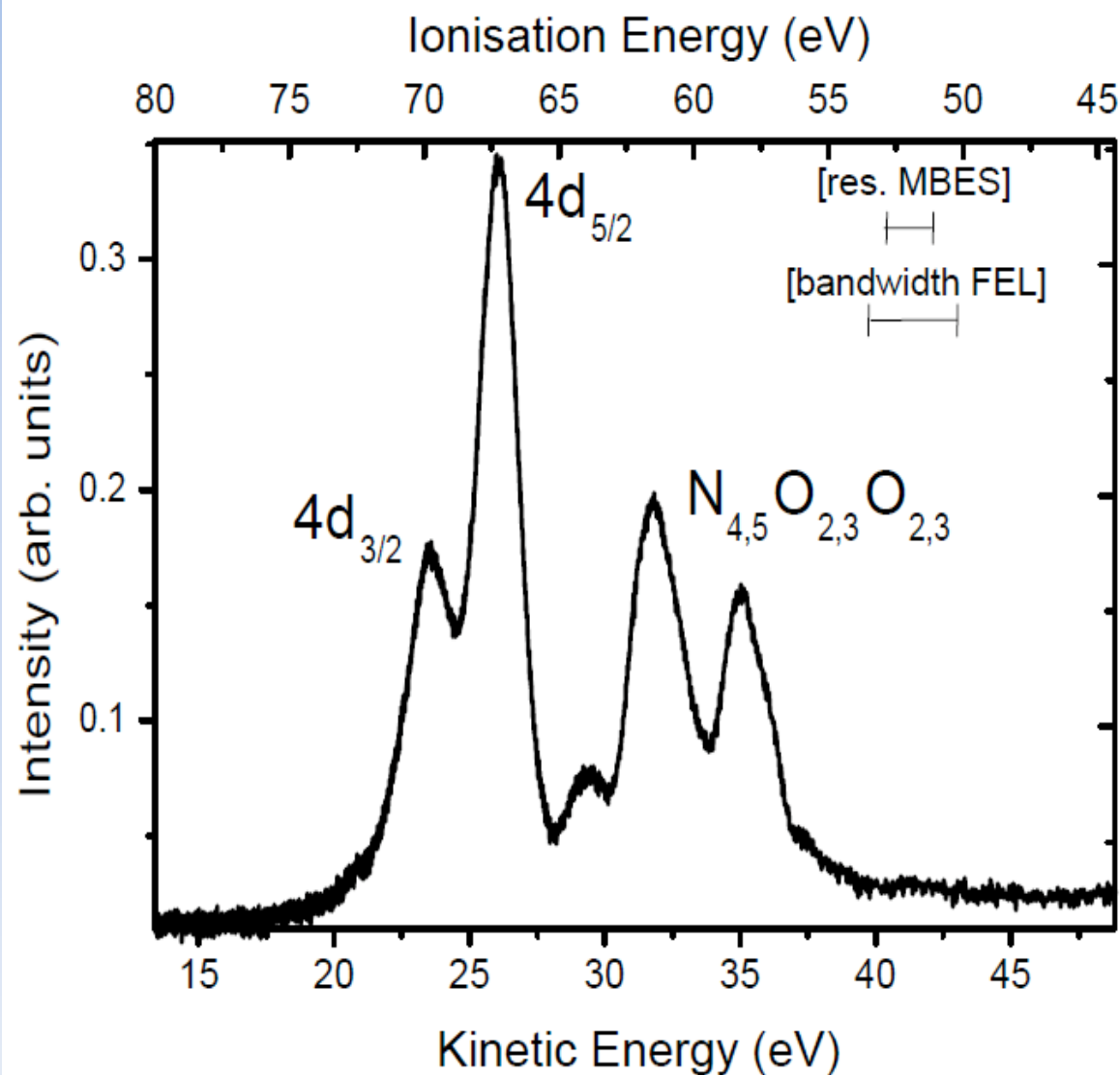
*Very highly charged ions observed - is MPI an important contributor ?*

Photoionization of xenon atoms in the EUV  
at ultra-high intensities: ion time-of-flight spectra



Sorokin, Richter et al., PTB, PRL 2007 –





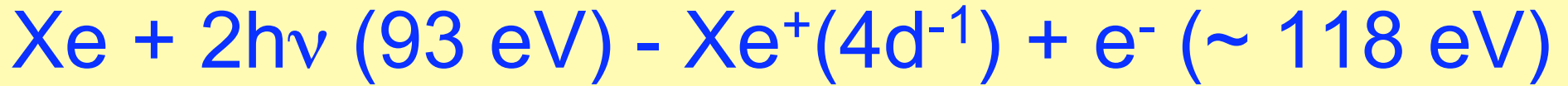
NB: We measure electrons and not ions !!!

- Single shot spectrum....

- For low intensities ( $<10^{13} \text{ W.cm}^{-2}$ ), one photon processes are dominant

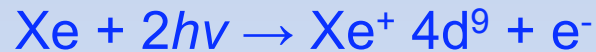
- Salient features – spin orbit split 4d photoelectron line + Auger electron spectrum

- Not shown –  $5s^{-1}$  and  $5p^{-1}$  lines at higher KEs



Now ramp up the intensity  
to  $> 10^{15} \text{ W.cm}^{-2}$ .....

- Using MBES, first evidence of two photon **inner** shell ionisation, (in this case) of 4d electron –

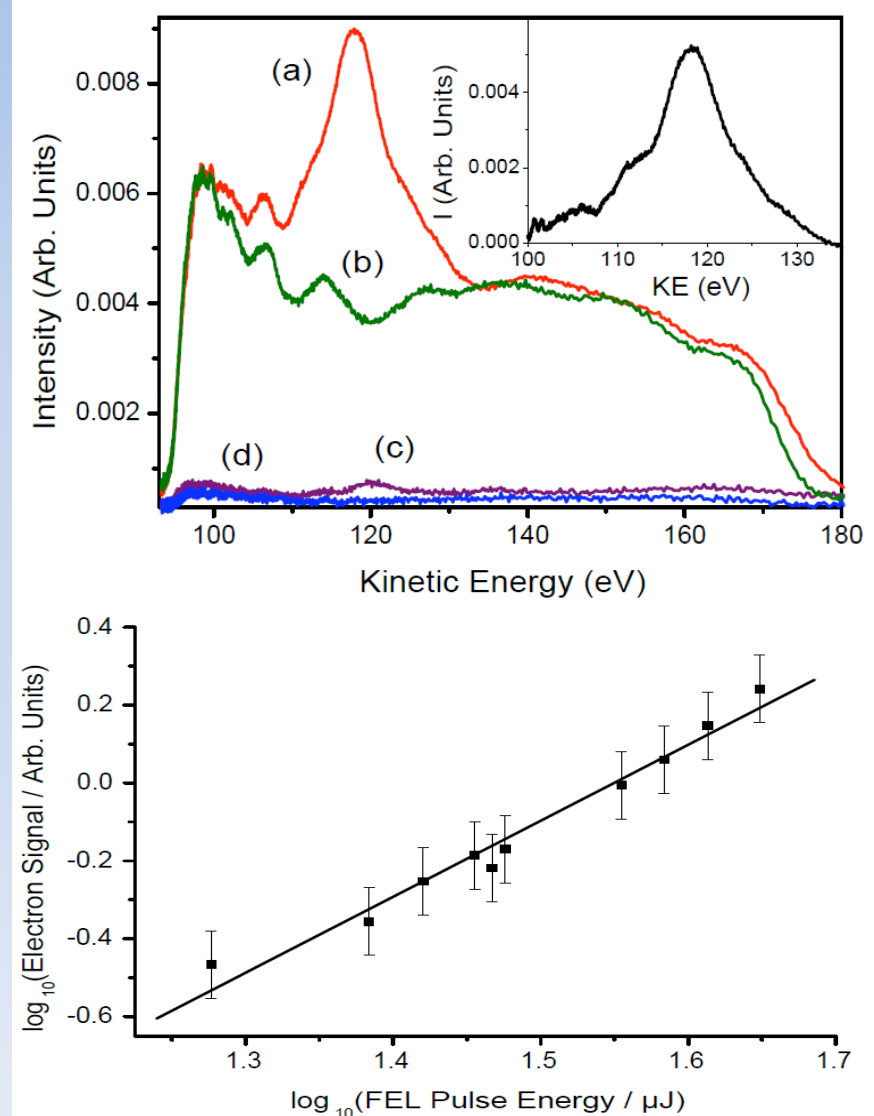


- ‘Retardation field’ applied to suppress low KE electrons (one photon processes) – hence electrons detected are due solely to multiphoton events

- Energetically –

$$2 \times (93) \text{ eV} - 118 \text{ eV} = 68 \text{ eV}$$

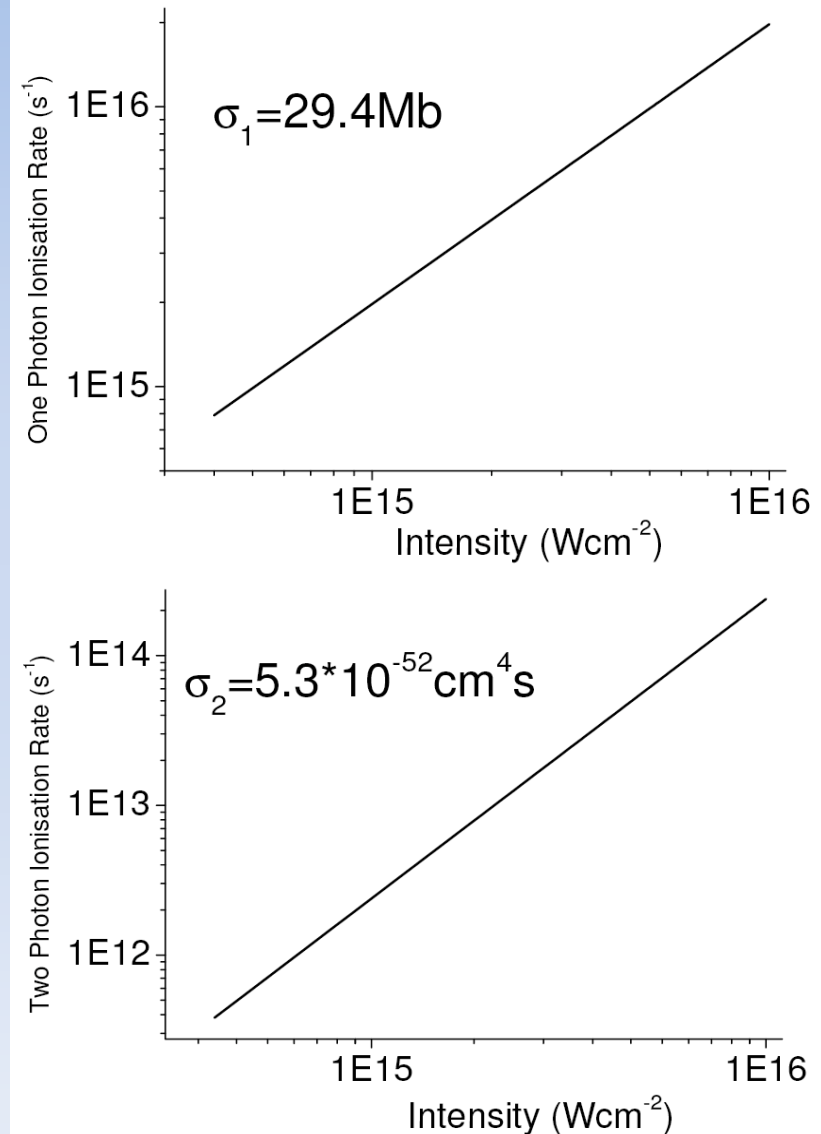
- Yield scales quadratically,  $n=1.95 \pm .$ ,  
 $\Rightarrow 2^{\text{nd}}$  Order Process !!





# 1 to 2 Photon Ionization Branching Ratio

1. R-Matrix (H.W. Van der Hart) – one and two photon 4d emission cross sections
2. *Dominant process is one photon ionization* – 93 eV can remove next 4d as well - or maybe excite 4p – 4d. High rate of ‘inside-out’ ionization.....
3. Accurate calculation requires a far more rigorous description of the atomic structure than at present
4. *Estimated two photon 4d<sup>-1</sup> emission is ~1% of total at ~ 7 x 10<sup>15</sup> W.cm<sup>-2</sup>*



# Read the full story here.....

PRL 105, 013001 (2010)

PHYSICAL REVIEW LETTERS

week ending  
2 JULY 2010

## Two-Photon Inner-Shell Ionization in the Extreme Ultraviolet

V. Richardson,<sup>1</sup> J. T. Costello,<sup>1</sup> D. Cubaynes,<sup>2</sup> S. Düsterer,<sup>3</sup> J. Feldhaus,<sup>3</sup> H. W. van der Hart,<sup>4</sup> P. Juranić,<sup>3</sup> W. B. Li,<sup>3,5</sup>  
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We have observed the simultaneous inner-shell absorption of two extreme-ultraviolet photons by a Xe atom in an experiment performed at the short-wavelength free electron laser facility FLASH. Photoelectron spectroscopy permitted us to unambiguously identify a feature resulting from the ionization of a single electron of the 4*d* subshell of Xe by two photons each of energy  $(93 \pm 1)$  eV. The feature's intensity has a quadratic dependence on the pulse energy. The results are discussed and interpreted within the framework of recent results of ion spectroscopy experiments of Xe obtained at ultrahigh irradiance in the extreme-ultraviolet regime.

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## Case 2. *Resonant* 2-photon Excitation (Kr)

1. To date we have looked at a **non-resonant** two photon process (sort of ATI really)
2. FELs are wavelength tunable - one can also explore **resonant two photon** processes

Kr  $3d^{10}4s^24p^6$  ( $^1S_0$ ) + 2 x  $h\nu$  (46 eV)  $\rightarrow$   $3d^94s^24p^64d$  (J=0,2)  
i.e., 3d - 4d two photon resonant excitation

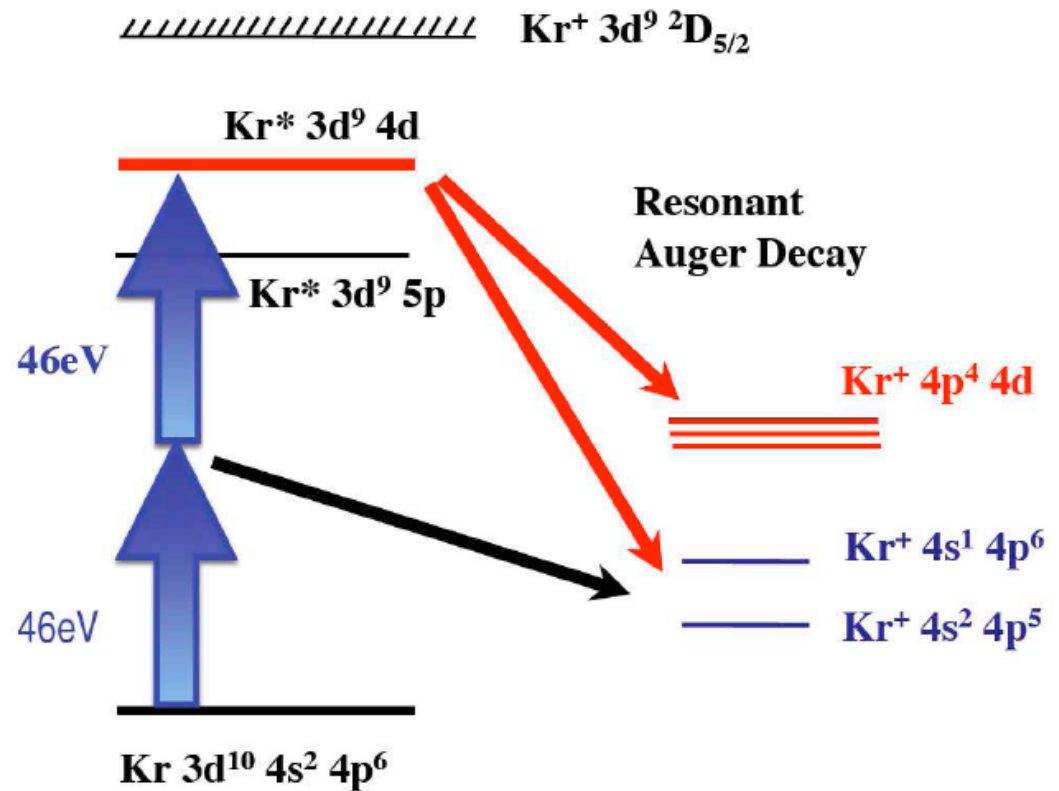
# Kr - Resonant Two Photon Excitation

1.  $\text{Kr } 3d^{10}4s^24p^6 (^1S_0) + 2 \times h\nu (46 \text{ eV}) \rightarrow 3d^94s^24p^64d (J=0,2)$  i.e., 3d - 4d two photon excitation

2. Of course there is a direct ionization path and the usual interference results - manifested as asymmetric resonance profiles (Fano/ Fano-Mies)

3. But here the  $3d^94s^24p^64d (J=0,2)$  resonance undergoes Auger decay to  $\text{Kr}^+$  on a femtosecond timescale - similar to the FLASH pulse duration - so competition between excitation and decay (ergo, in addition to simple ATI, this case makes for an intriguing, problem for theory)..

$$h\nu = 46 \text{ eV } (\sim 27 \text{ nm})$$

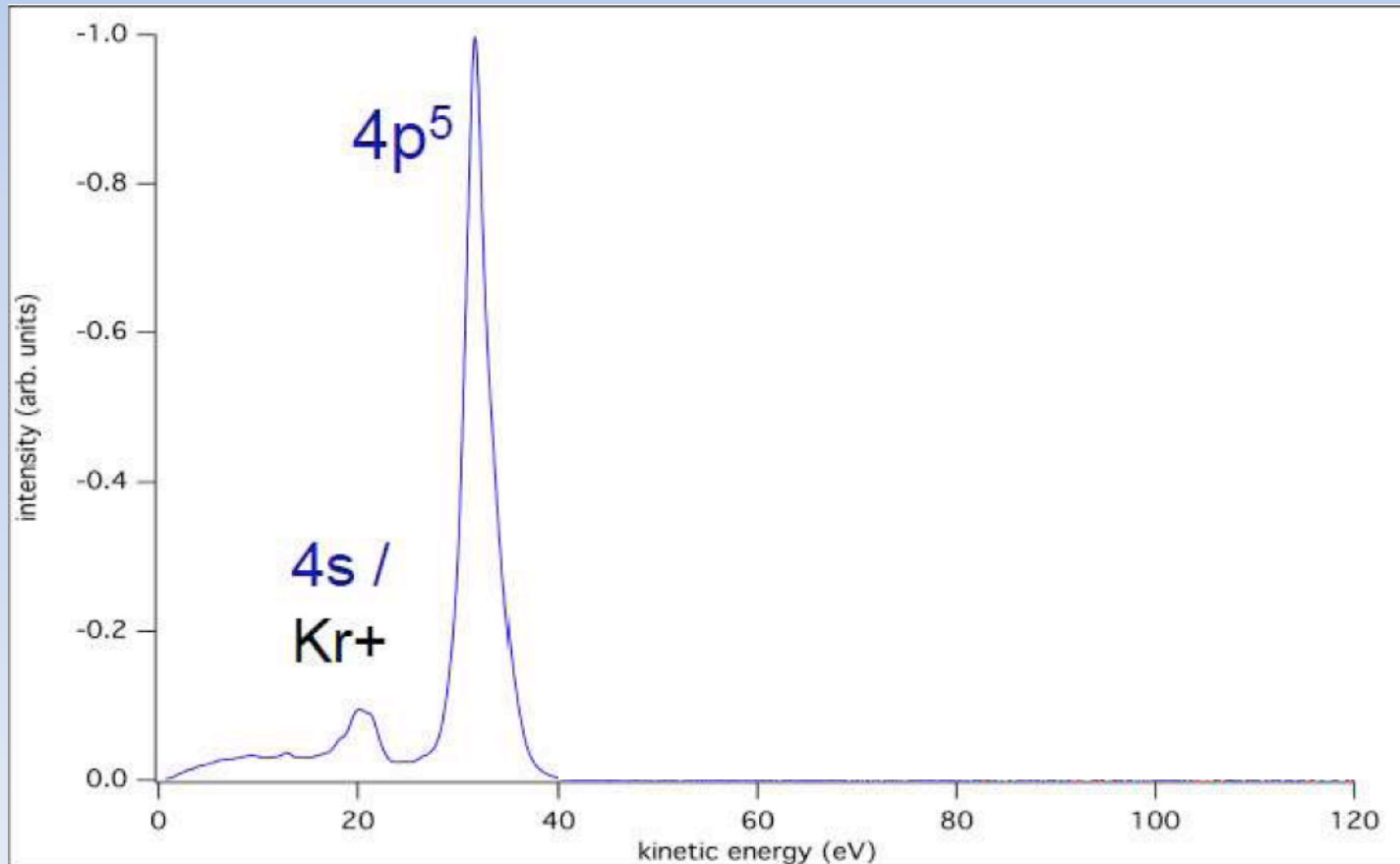


Meyer et al., PRL **104** 213001 (2010)



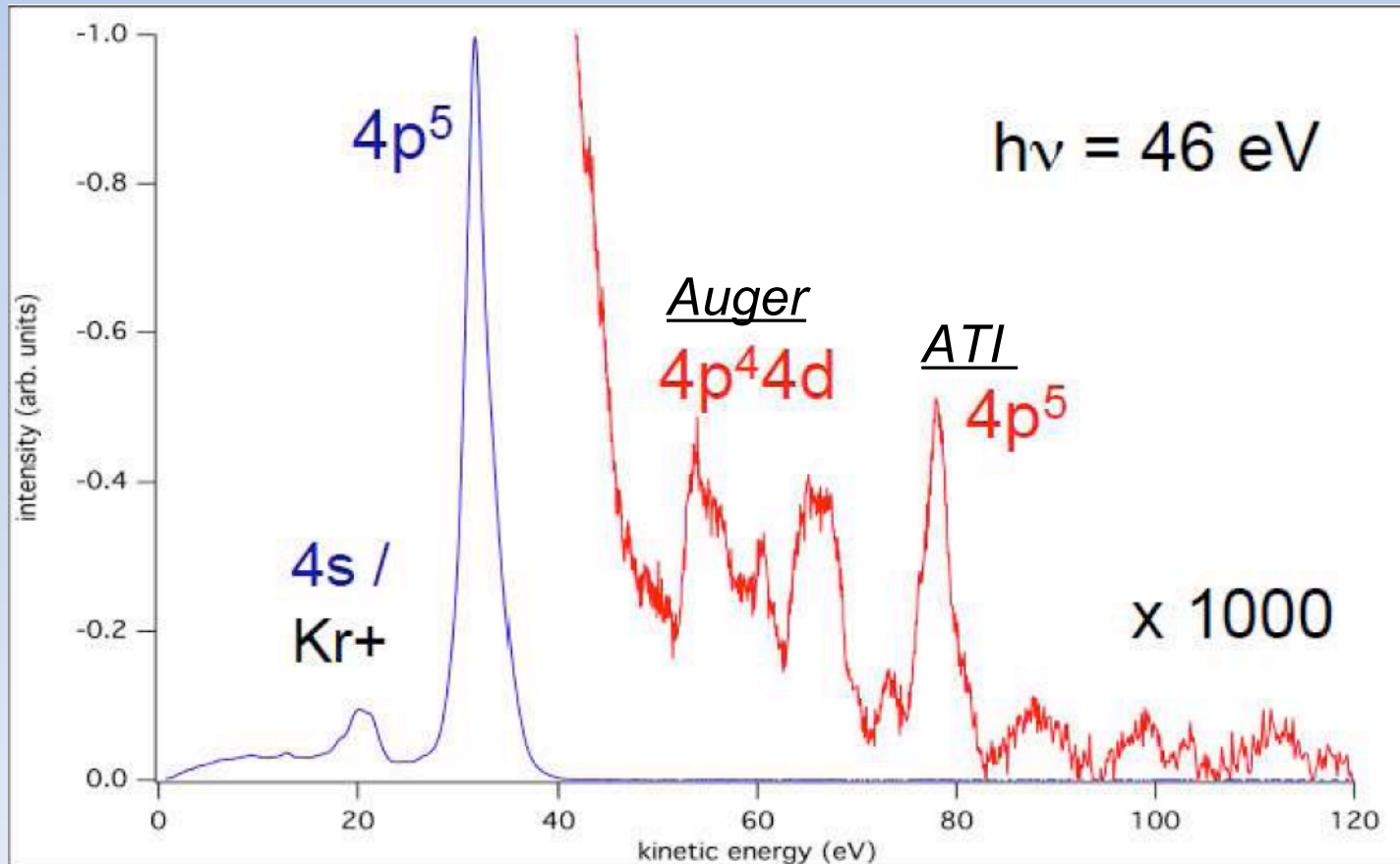
# Kr (3d<sup>9</sup>4d) 2 Photon Resonant Auger

MBES Photoelectron spectrum -  $\sim 10^{14}$  W.cm<sup>-2</sup>



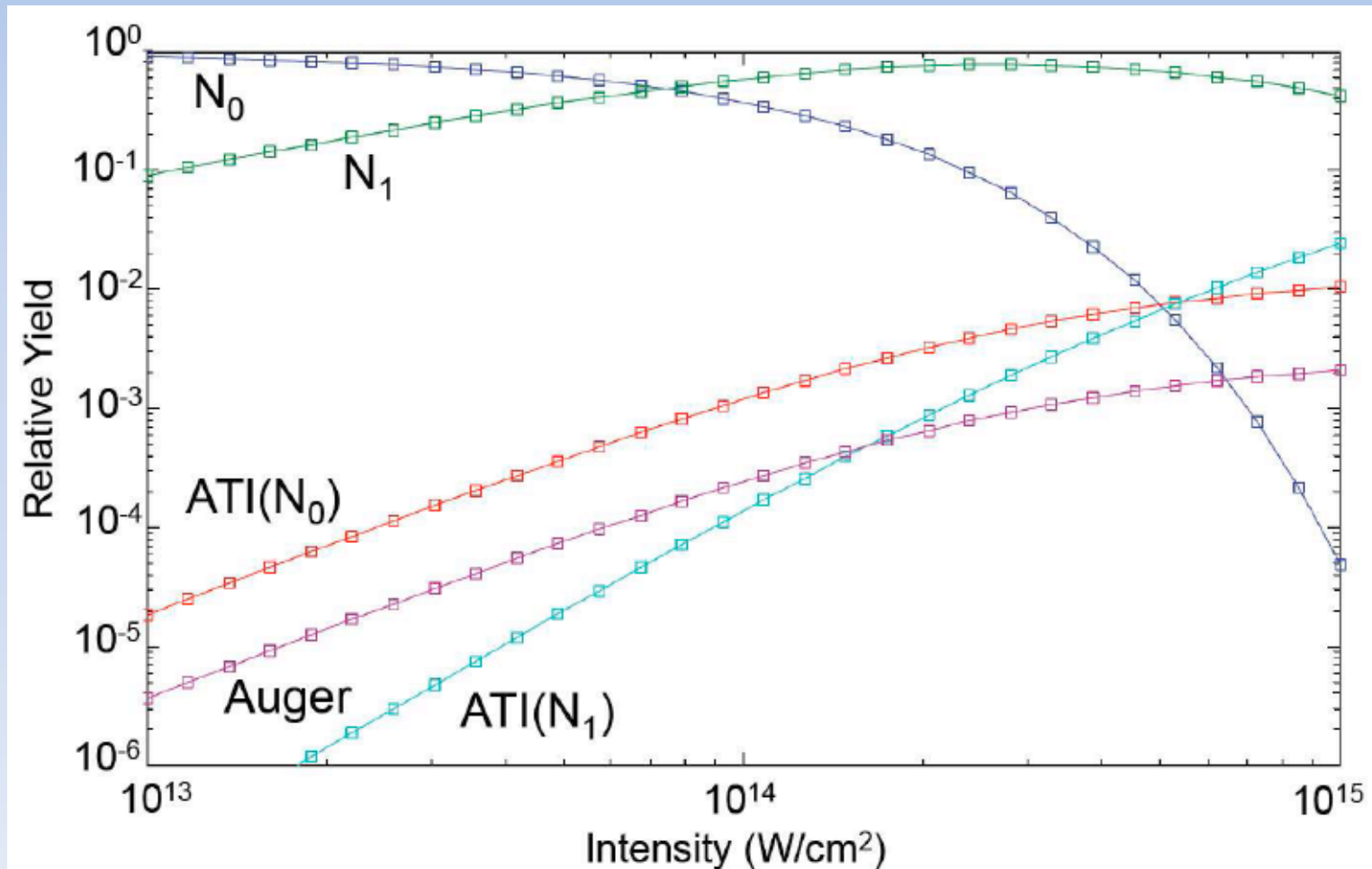
# Kr ( $3d^94d$ ) 2 Photon Resonant Auger

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# Kr (3d<sup>9</sup>4d) 2 Photon Resonant Auger

Ionization rates – P. Lambropoulos, Crete



# Read more here.....

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PHYSICAL REVIEW LETTERS

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## Two-Photon Excitation and Relaxation of the $3d$ - $4d$ Resonance in Atomic Kr

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Two-photon excitation of a single-photon forbidden Auger resonance has been observed and investigated using the intense extreme ultraviolet radiation from the free electron laser in Hamburg. At the wavelength 26.9 nm (46 eV) two photons promoted a  $3d$  core electron to the outer  $4d$  shell. The subsequent Auger decay, as well as several nonlinear above threshold ionization processes, were studied by electron spectroscopy. The experimental data are in excellent agreement with theoretical predictions and analysis of the underlying multiphoton processes.

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# Summary - One Colour

- Xenon - First detection of a so-called 'above threshold ionization' (ATI) two-photon process in an *inner shell electron* shell.
- It is clear that although single photon ionization processes dominate, they are sufficiently important at high irradiance that, for a given intensity, much higher ionization stages can be reached compared to optical lasers.
- The strength and the nature of the  $4d \rightarrow \epsilon f$  resonance may open up, at high irradiance, additional ionization channels, namely the *simultaneous multiphoton / multi-electron from the inner 4d shell*, '*inside-out ionization*' or '*peeling the onion from the inside out*'
- *Kr - first step on the road to resonant NL processes with EUV/X-rays.... REMPI at X-ray.*

*Xe - Richardson et al. PRL (July 2 – 2010), Kr - Meyer et al., PRL (May 28 - 2010)*

## 5. Summary and Next Steps

1. We have demonstrated interference free *sidebands (SB) to high order*, SB polarisation *control*, laser & FEL dependencies & SBs in atoms/ions
2. Especially useful as an *EUV-Optical cross correlation technique*
3. First resonant and non-resonant *two photon ionization processes involving inner-shell electrons demonstrated at FLASH*
4. Experiments started at LCLS (summer 2009) - SBs have been used to measure 'long' (> 100 fs) pulse widths
5. First few fs streak measurements at LCLS (July 2010) - under analysis
6. First data on *angular distribution of SB electrons\** at LCLS (last week)
7. LCLS will test the limits of XF X-ray pulse width and X-ray optical jitter measurement techniques – 'Tandem streaking' idea of Adrian Cavalieri just accepted for beamtime.....
8. Next step (physics) - X-ray laser pumping, coherent control, Rabi flops,...
9. 2011 and beyond: FLASH - *seeding (sFLASH)*, LCLS/XFEL - *aluminium 'slotted spoiler' for cleaner pulses, seeding (Echo 7 project)*,.....

\* cf: Kazansky AK, Kabachnik NM, JOURNAL OF PHYSICS B-AMOP, **42** Article No. 121002 (2009)



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